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(54) ULTRAFILTRATION MEMBRANE, HYDROGEN SEPARATION MEMBRANE, METHOD FOR
MANUFACTURING THE MEMBRANE, AND METHOD FOR SEPARATING HYDROGEN

(57)Abstract:

PROBLEM TO BE SOLVED: To provide an ultrafiltration membrane which has heat stability and chemical resistance, is resistant to physical impact, excellent in high temperature connectivity, and useful as a hydrogen separation membrane.

SOLUTION: In the ultrafiltration membrane, a void part of a membrane formed of a sintered metal is filled with fine metal particles and/or fine metal oxide particles and/or fine ceramic particles.

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CLAIMS

[Claim(s)]

[Claim 1]

Ultrafiltration membrane which fills up with metal particles, a metal oxide particle, and/or ceramic particles a cavity part of a film formed with a sintered metal, and is characterized by things.

[Claim 2]

Hydrogen separation membrane having used as a base material a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal, and sticking palladium membrane on it.

[Claim 3]

Hydrogen separation membrane having used as a base material a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal, and sticking (b) palladium membrane and/or a thin film of a (**) palladium alloy on it.

[Claim 4]

The hydrogen separation membrane according to claim 3 being the precious metals beyond a kind as which a palladium alloy is chosen from a group which consists of palladium, silver and gold, platinum, nickel, and cobalt, or a NI kind.

[Claim 5]

A manufacturing method of the ultrafiltration membrane according to claim 1 circulating metal particles, a metal oxide particle, and ceramic particles in an opening of a film formed with a sintered metal.

[Claim 6]

A manufacturing method of the hydrogen separation membrane according to claim 2 using as a base material a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal, and sticking palladium membrane on it.

[Claim 7]

A film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, A manufacturing method of the hydrogen separation membrane according to claim 3 sticking (b) palladium membrane and/or a thin film of a palladium alloy on it.

[Claim 8]

A separation method of hydrogen using the hydrogen separation membrane according to claim 2 which used as a base material a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal, and stuck palladium membrane on it.

[Claim 9]

A film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, A separation method of hydrogen using the hydrogen separation membrane according to claim 3 to which (b) palladium membrane and/or a thin film of a (**) palladium alloy were stuck on it.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]**[0001]****[Field of the Invention]**

It is related with the separation method of the hydrogen which uses the ultrafiltration membrane which this invention fills up with metal particles, a metal oxide particle, and/or ceramic particles the cavity part of the film formed with the sintered metal, and is characterized by things, its manufacturing method, and it.

[0002]**[Description of the Prior Art]**

TK film from which an effective pole diameter is constituted by fine pores (0.01 microns or more and 0.2 micron or less) is called ultrafiltration membrane, and has a use as a structure of the transmission body of separation uses, such as separation of a gaseous mixture, vapor liquid separation, and solid liquid separation, catalyst support, a gas or gases, such as a base material of a liquid separation film, and/or a fluid, etc. As such a substance, porous-ceramics films, such as polymers porous membrane, porous glass films, and alumina, a sintered metal film, etc. are mentioned. However, a polymer material is lacking in heat and chemical tolerance, and porous glass films are usually deficient in tolerance to a steam. When the junction nature to the metal of what those faults do not have uses porous ceramics, such as alumina, at an elevated temperature especially bad, the connection with a device is not easy. In that respect, although the sintered metal film is excellent in such heat, chemical tolerance, and connectivity, it is difficult to make an effective pole diameter small, and an effective pole diameter is usually 0.2 microns or more. Then, by applying to such the sintered metal film surface the ceramic solution suspended, for example, an effective pole diameter coats 0.01 to 0.2 micron of ceramics membrane, and considers it as ultrafiltration membrane. However, by such a film, since the thermal expansion nature of ceramics and metal differs, when a thermal shock is received, the junction nature of metal-ceramics gets worse, the crack of ceramics membrane, the exfoliation from the sintered metal film surface, etc. arise as the result, and the performance degradation in an elevated temperature is brought about sometimes. There is a physical shock, for example, a possibility that the ceramics membrane shown in an outside surface by contact with the particles in a penetration fluid may get damaged, and performance may deteriorate. Although such ultrafiltration membrane is used as a base material of palladium membrane or a palladium alloy thin film and it is used as hydrogen separation membrane, since the same problem arises too, industrial utility value is spoiled.

[0003]**[Problem(s) to be Solved by the Invention]**

This invention solves the above-mentioned problem, has thermal stability and chemical tolerance, is strong also against a physical shock and provides the extra demarcation membrane and hydrogen separation membrane which were moreover excellent also in hot connectivity.

[0004]**[Means for Solving the Problem]**

A result to which this invention person advanced various experiment and researches to such a technical problem, If an opening of a sintered metal film is filled up with metal particles, a metal oxide particle, and/or ceramic particles, 0.2 micron or less of ultrafiltration membrane will be obtained for an effective pole diameter, if it finds out that it becomes easy to acquire an expected effect and palladium membrane or a palladium alloy thin film is formed on it by using this ultrafiltration membrane as a base material, acquiring an expected effect will find out becoming easy and it will come to complete this invention. A manufacturing method of these films is very easy, and manufacture of a **** large quantity is also easy for it in a large-scale manufacturing facility.

[0005]

Namely, this invention

- (1) Ultrafiltration membrane which fills up with metal particles, a metal oxide particle, and/or ceramic particles a

cavity part of a film formed with a sintered metal, and is characterized by things,

(2) Hydrogen separation membrane having used as a base material a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal, and sticking palladium membrane on it,

(3) A film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, Hydrogen separation membrane sticking (b) palladium membrane and/or a thin film of a (**) palladium alloy on it,

(4) Hydrogen separation membrane given in the above (3) being the precious metals beyond a kind as which a palladium alloy is chosen from a group which consists of palladium, silver and gold, platinum, nickel, and cobalt, or a Ni kind,

(5) A manufacturing method of ultrafiltration membrane given in the above (1) circulating metal particles, a metal oxide particle, and ceramic particles in an opening of a film formed with a sintered metal,

(6) a cavity part of a film formed with a sintered metal -- (**) -- metal particles, a metal oxide particle, and/or (**) -- a manufacturing method of hydrogen separation membrane given in the above (2) using as a base material a film filled up with ceramic particles, and sticking palladium membrane on it,

(7) A film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, moreover -- (**) -- a manufacturing method of hydrogen separation membrane given in the above (3) sticking palladium membrane and/or a thin film of a palladium alloy,

(8) a film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, A separation method of hydrogen using hydrogen separation membrane of a statement for the above (2) to which palladium membrane was stuck on it,

(9) A film which fills up with (b) metal particles, a metal oxide particle, and/or (**) ceramic particles a cavity part of a film formed with a sintered metal is used as a base material, moreover -- (**) -- palladium membrane and/or (**) -- a separation method of hydrogen using hydrogen separation membrane of a statement for the above (3) to which a thin film of a palladium alloy was stuck,

It is alike and is related.

[0006]

[Embodiment of the Invention]

The invention concerning claim 1

In the ultrafiltration membrane of this invention, it is desirable to use a sintered metal film as a base material, and to fill up this base material opening with metal particles, a metal oxide particle, and/or ceramic particles.

As construction material of a sintered metal film, stainless steel, the Hastelloy alloy, Inconel, nickel, a nickel alloy, titanium, a titanium alloy, etc. are mentioned. Although there is no restriction in particular in the effective pore diameter of a sintered metal film, it is preferred that an effective pore diameter is about 0.2 micron.

However, even if the effective pore diameter is 0.2 microns or more, so that claim 2 may describe, Since the effective pore diameter can be about 0.2 micron by circulating the solution having contained the metal particles, metal oxide particle, and/or ceramic particles of grain size which suit the effective pore diameter in fine pores, it is convenient. For example, with metal plating, such as nickel, copper, chromium, zinc, tin, and iron, the inside of the sintered metal film surface and fine pores may be embellished with metal, and an effective pore diameter may be made small. Especially if nickel, iron, silver, copper, palladium, etc. are metal which is particles as metal particles with which a sintered metal film opening is filled up, it will not be restricted, but it is indispensable that the particles below the effective pore diameter of the film from which the effective particle diameter serves as a base material are contained. As the metal oxide particle with which a sintered metal film opening is filled up and/or ceramics, for example, An aluminum oxide, iron oxide, zirconium oxide, cerium oxide, silver oxide, Especially if copper oxide, oxidation aluminum, zirconium oxide, silicon nitride, etc. are metal oxide or ceramics which are particles, it will not be restricted, but as for the effective particle diameter, it is indispensable that it is below the effective pore diameter of the film used as a base material. When gas permeation is performed using such ultrafiltration membrane, for example it is based on the transmission rate of argon gas, the transmission rate of hydrogen will be the 3.5 or more times. This is for the Knudsen style to arise and for the difference in a transmission rate to arise by the size of the molecular weight of penetration gas, if an effective pore diameter arrives at a 0.01 to 0.2-micron field. The degree of restoration of the particles to the inside of the opening of a sintered metal film will become clear easily, if a membranous section is observed using a scanning electron microscope. When it is such a structure, the fine pores which serve as a gestalt with which particles are held and the particle forms all over the opening of sintered metal achieve the function as ultrafiltration membrane. Therefore, though a filtration membrane outside surface is damaged, degradation of big performance is not produced.

[0007]

The invention concerning claim 2

It is preferred to form palladium membrane on it by using as a base material ultrafiltration membrane indicated to claim 1 in the hydrogen separation membrane of this invention. It is known very well that palladium membrane has alternative penetrability of hydrogen. Although palladium membrane can be formed with sufficient adhesion on a sintered metal film by using as a base material ultrafiltration membrane indicated to claim 1, For example, a lot of [since the effective pole diameter on a sintered metal film is too large when palladium membrane is formed on a mere sintered metal film, in order to close fine pores by palladium thoroughly] palladium is needed, And since the opening of a sintered metal film exists, it results in adhesion with palladium membrane being spoiled and losing membranous stability.

As palladium membrane thickness on ultrafiltration membrane, 50 microns is usually 20 microns from 5 microns preferably from 3 microns. If thickness is too small, the preferential segregation performance of hydrogen will be lost, and economical efficiency will be lost if thickness is too large.

[0008]

The invention concerning claims 3 and 4

It is preferred to coat palladium membrane and/or the thin film of a palladium alloy on it by using as a base material ultrafiltration membrane indicated to claim 1 in the hydrogen separation membrane of this invention. The metal which constitutes a palladium alloy is a kind chosen from the group which consists of palladium, silver and gold, platinum, nickel, cobalt, etc., or two sorts or more (invention concerning claim 4.). The following is also the same. Such a palladium alloy thin film has alternative penetrability of hydrogen, and it is known very well that hydrogen permeation performance will increase by alloying.

As palladium-alloy-membrane thickness on ultrafiltration membrane, 50 microns is usually 20 microns from 5 microns preferably from 3 microns. If thickness is too small, the preferential segregation performance of hydrogen will be lost, and economical efficiency will be lost if thickness is too large.

[0009]

The invention concerning claim 5

This invention is a manufacturing method of the ultrafiltration membrane indicated to above-mentioned claim 1, These particles are fixed in the opening of a sintered metal film by making metal particles, a metal oxide particle, ceramic particles, and/or metal hydroxide particles suspended to a solvent, and making a sintered metal film penetrate it. As for especially the effective particle size of particles, for the increase in efficiency of the thing deposition which is not restricted, since it is necessary to deposit and fix particles all over the opening of a sintered metal film in the invention concerned, it is preferred to carry out an effective particle size near the effective pole diameter. If too large, it will not go into fine pores, and if too small, fine pores are passed and it is not fixed all over an opening. About a presentation, there are no restrictions only by the particle diameter having restriction as particles. Since the inorganic substance which removes an organic matter by operation of the calcination in the air, etc., and constitutes particles will remain all over the opening of a sintered metal film even if organic matter has adhered to particles for example, it is satisfactory. Even if it is the particles by which metal hydroxide adhered to ceramic particles, it will be satisfactory if the effective particle size is near the effective pole diameter of the opening in a sintered metal film.

What is necessary is just to perform the penetration to the film of particle suspension by pressing liquid fit in fine pores from field of membranous one of the two, or making membranous one of the two decompression, and attracting liquid. By making a sintered metal film pass the particles which have an effective particle size according to the pole diameter, even if the effective pole diameter of a sintered metal film is 0.2 microns or more, An effective pole diameter can obtain 0.2 micron or less of ultrafiltration membrane by repeating the same operation, deposition all over the opening of the sintered metal film of this particle arising, and an effective pole diameter becoming small, and making the effective diameter of particles small after that. A better result will be obtained if shake of the liquid by an ultrasonic wave etc. and a sintered metal film is performed in the case of the penetration operation to the film of this particle suspension.

After performing deposition all over the sintered metal film opening of particles, even if the particle diameter of particles is changed, particle suspension is penetrated succeeding and it performs further control of an effective pole diameter, there is no inconvenience, but. Stabilization into the opening of particles is once attained by drying or calcinating by operation of heating etc. after deposition of particles, The liquid suspended in particles on this film can be passed, and the ultrafiltration membrane which has a desired effective pole diameter can be obtained by repeating the operation which fixes particles into the opening which remains. Although the effective particle size in particular of the particles used at this time is not limited, for the increase in efficiency of deposition, it is preferred that it is near the effective pole diameter of the fine pores which the opening which remains forms.

[0010]

The invention concerning claim 6

This invention is a manufacturing method of the hydrogen separation membrane indicated to above-mentioned claim 2, by using the ultrafiltration membrane according to claim 1 as a base material, on it, is publicly known, for example, should just form palladium membrane with palladium membrane preparation methods, such as a nonelectrolytic plating method, the electrolysis plating method, and chemical vapor deposition. Even if it uses ultrafiltration membrane immediately after making particles deposit, it is convenient. If it carries out by in the palladium membrane formation by the electrolysis plating method or nonelectrolytic plating pressing plating liquid fit in fine pores from the field of one of the two of ultrafiltration membrane, or making decompression one of the two of ultrafiltration membrane, and attracting liquid, the adhesion of palladium membrane will increase.

[0011]

The invention concerning claim 7

This invention is a manufacturing method of the hydrogen separation membrane indicated to above-mentioned claim 3, and should just form palladium membrane and/or a palladium alloy thin film above-mentioned on it by a publicly known method by using the ultrafiltration membrane according to claim 1 as a base material. Forming a palladium alloy thin film directly on ultrafiltration membrane as an alloy thin film preparation method, for example with publicly known palladium alloy thin film preparation methods, such as the electrolysis plating method, a nonelectrolytic plating method, and chemical vapor deposition, is mentioned. Otherwise, for example with palladium membrane preparation methods, such as the electrolysis plating method, a nonelectrolytic plating method, and chemical vapor deposition, after forming palladium membrane on ultrafiltration membrane, Heating the precious metals, such as silver, gold, platinum, nickel, and cobalt, in reducing gas, such as hydrogen, at the temperature of not less than 300 ** after coating the surface with a publicly known method, for example, the electrolysis plating method, a nonelectrolytic plating method, or chemical vapor deposition is mentioned. These may be performed in accordance with the publicly known method.

[0012]

The invention concerning claim 8

This invention is a separation method of the hydrogen which uses the hydrogen separation membrane indicated to above-mentioned claim 2. If the gas containing hydrogen is put on one side (hydrogen containing gas side) of the hydrogen separation membrane indicated to claim 2 as a separation method of hydrogen and hydrogen content pressure of the opposite hand (hydrogen side) is made below into the hydrogen content pressure by the side of hydrogen containing gas, Hydrogen can penetrate the inside of hydrogen separation membrane selectively, and hydrogen in the hydrogen containing gas side can be divided into the hydrogen side. 700 ** of this hydrogen separation membrane can usually be preferably used at the temperature of 300 to 600 ** from 200 **. If temperature is too low, embrittlement of palladium membrane will arise, and if temperature is too high, degradation of palladium membrane will arise. If such hydrogen separation membrane is used, the selective separation of hydrogen will become possible.

[0013]

The invention concerning claim 9

This invention is a separation method of the hydrogen which uses the hydrogen separation membrane indicated to above-mentioned claim 3. If the gas containing hydrogen is put on one side (hydrogen containing gas side) of the hydrogen separation membrane indicated to claim 2 as a separation method of hydrogen and hydrogen content pressure of the opposite hand (hydrogen side) is made below into the hydrogen content pressure by the side of hydrogen containing gas, Hydrogen can penetrate the inside of hydrogen separation membrane selectively, and hydrogen in the hydrogen containing gas side can be divided into the hydrogen side. 700 ** of this hydrogen separation membrane can usually be preferably used at the temperature of 300 to 600 ** from 200 **. If temperature is too low, embrittlement of palladium alloy membrane will arise, and if temperature is too high, degradation of palladium alloy membrane will arise. If such hydrogen separation membrane is used, the selective separation of hydrogen will become possible.

[0014]

[Example]

The example of manufacture

The example of manufacture of ultrafiltration membrane and hydrogen separation membrane is given to below, and let the place by which it is characterized [of this invention] be a further still clearer thing.

[0015]

The example 1 of manufacture

It put into the solution which made the zirconium hydroxide sol to which an effective particle diameter is distributed over 1.0 micron from 0.07 micron in the sintered-metal pipe made from stainless steel whose

effective pole diameter which stopped one side is 0.2 micron suspended, and the solution which decompressed the opening of the metal tube and made zirconium hydroxide sol suspended was attracted. At this time, shake by an ultrasonic wave was performed and distribution of zirconium hydroxide sol and diffusion of the zirconium hydroxide sol to the opening of a sintered metal film were made easy. After drawing in for 3 hours, it dried at 120 °C for 5 hours, and heated in the air at 400 °C after that for 2 hours. In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the obtained ultrafiltration membrane was 135 ml/cm²/min, and argon permeation speed was 37 ml/cm²/min (H₂/Ar=3.6). In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the sintered-metal pipe used as a base material was 344 ml/cm²/min, and argon permeation speed was 131 ml/cm²/min (H₂/Ar=2.6).

[0016]

The example 2 of manufacture

Ultrafiltration membrane was obtained using the solution which made the zirconium hydroxide sol which used as the base material ultrafiltration membrane obtained in the example 1 of manufacture, and was distributed over 0.1 micron from 0.05 micron by the same technique as the example 1 of manufacture suspended. In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the obtained ultrafiltration membrane was 58 ml/cm²/min, and argon permeation speed was 16 ml/cm²/min (H₂/Ar=4.1).

[0017]

The example 3 of manufacture

Ultrafiltration membrane was obtained using the solution which made the nickel particulates which used as the base material ultrafiltration membrane obtained in the example 1 of manufacture, and were distributed over 0.8 micron from 0.05 micron by the same technique as the example 1 of manufacture suspended. In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the obtained ultrafiltration membrane was 67 ml/cm²/min, and argon permeation speed was 18 ml/cm²/min (H₂/Ar=3.7).

[0018]

The example 4 of manufacture

Ultrafiltration membrane was obtained using the solution which made the hydroxylation palladium zirconium hydroxide coprecipitation neutralized precipitate which used as the base material ultrafiltration membrane obtained in the example 1 of manufacture, and was distributed over 0.8 micron from 0.1 micron by the same technique as the example 1 of manufacture suspended. In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the obtained ultrafiltration membrane was 41 ml/cm²/min, and argon permeation speed was 10 ml/cm²/min (H₂/Ar=4.1).

[0019]

The example 5 of manufacture

It put into the solution which made the nickel particulates to which an effective particle diameter is distributed over 2 microns from 0.07 micron in the sintered-metal pipe made from stainless steel whose effective pole diameter which stopped one side is 1 micron suspended, the opening of the metal tube was decompressed, and the solution which made nickel particulates suspended was attracted. Then, this metal tube was put into the solution which made zirconium hydroxide sol suspended, the opening of the metal tube was decompressed, and the solution which made zirconium hydroxide sol suspended was attracted. At this time, shake by an ultrasonic wave was performed and distribution of water cerium oxide sol and diffusion of the zirconium hydroxide sol to the opening of a sintered metal film were made easy. After drawing in for 3 hours, it dried at 120 °C for 5 hours, and heated in the air at 400 °C after that for 2 hours. In differential pressure 0.2atm, the hydrogen permeation speed per unit membrane area of the obtained ultrafiltration membrane was 30 ml/cm²/min, and argon permeation speed was 8 ml/cm²/min (H₂/Ar=3.8).

[0020]

The example 6 of manufacture

It was immersed in the alkaline catalyst grant solution of marketing of the ultrafiltration membrane obtained in the example 3 of manufacture at the room temperature, and the palladium core was formed in the ultrafiltration membrane surface. It was immersed in the unelectrolyzed palladium plating liquid of marketing of this at 60 °C, the opening of the metal tube was decompressed, and plating liquid was attracted. When the flow of plating liquid stopped, ultrafiltration membrane was pulled up from plating liquid, and it washed and dried. The palladium membrane thickness which became clear by scanning electron microscope observation was 7 - 10 microns after hydrogen separation system performance testing.

[0021]

The example 7 of manufacture

The stainless steel sintered-metal pipe whose effective pore diameter which stopped one side is 0.5 micron was put into the commercial nickel strike plating bath, electroplating was performed, and the nickel thin film was formed in the inside of a sintered-metal pipe surface and fine pores. the hydroxylation cerium distributed over 3 microns from 0.2 micron in this sintered-metal pipe -- putting into the solution which made sol suspended and decompressing the opening of a metal tube -- hydroxylation cerium -- sol was attracted. Then, it put into the solution which made the nickel particulates distributed over 0.5 micron from 0.1 micron in this metal tube suspended, the opening of the metal tube was decompressed, and nickel particulates were attracted. Thus, it washed and dried, after putting the obtained ultrafiltration membrane into the palladium silver alloy plating bath constituted with a palladium chloride, silver sulfate, EDTA, ethylenediamine, ammonium carbonate, and an ammonia solution and carrying out electrolysis plating of the palladium silver alloy thin film. The palladium-alloy-membrane thickness which became clear by scanning electron microscope observation was 10-15 microns after hydrogen separation system performance testing.

[0022]

The example of a hydrogen separation method is given to below, and let the place by which it is characterized [of this invention] be a further still clearer thing.

[0023]

Example 1

The hydrogen separation membrane obtained in the example 6 of manufacture was kept at 300 **, the gaseous mixture of 1.4 atmospheres of hydrogen and 1.4 atmospheres of argon was put on the outside of a demarcation membrane, and the inside of hydrogen separation membrane was made into atmospheric pressure. As a result, hydrogen was separated inside from the membranous outside by the flow of 7 ml/cm²/min. There was no penetration of argon.

[0024]

Example 2

The hydrogen separation membrane obtained in the example 6 of manufacture was kept at 600 **, the gaseous mixture of 1.4 atmospheres of hydrogen and 1.4 atmospheres of argon was put on the outside of a demarcation membrane, and the inside of hydrogen separation membrane was made into atmospheric pressure. As a result, hydrogen was separated inside from the membranous outside by the flow of 22 ml/cm²/min. There was no penetration of argon.

[0025]

Example 3

The hydrogen separation membrane obtained in the example 7 of manufacture was kept at 500 **, the gaseous mixture of 1.4 atmospheres of hydrogen and 1.4 atmospheres of argon was put on the outside of a demarcation membrane, and the inside of hydrogen separation membrane was made into atmospheric pressure. As a result, hydrogen was separated inside from the membranous outside by the flow of 15 ml/cm²/min. There was no penetration of argon.

[0026]

[Effect of the Invention]

As compared with a publicly known extra demarcation membrane, the ultrafiltration membrane by this invention is thermally and mechanically stable, and, moreover, does not require complicated operation of manufacture. Therefore, use to catalytic reaction hot [accompanied by rough separation of a hot gas, the filtering separation of a solid and a fluid, and gas permeation] is possible. As compared with publicly known hydrogen separation membrane, the hydrogen separation membrane by this invention is thermally and mechanically stable, and, moreover, does not require complicated operation of manufacture. Therefore, it can be used for the selective separation of hot hydrogen.

[Translation done.]